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## A new flowsheeting tool for flue gas treating

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### Abstract

A new flowsheeting tool, specifically designed for steady-state simulation of acid gas treating processes, has been developed. The models implemented in the new tool combine all issues relevant for the design, optimization and analysis of acid gas treating processes, including post-combustion and pre-combustion carbon dioxide capture. The computer code consists of an extremely user-friendly graphical user interface and a very powerful numerical simulator that handles rigorous modeling of thermodynamics, activity based kinetics, rate-based mass transfer and supports all unit operations relevant for gas treating plants (absorbers, strippers, flash drums, heaters, pumps, compressors, mixers and splitters, etc.).

Although the simulator can be used as a multifunctional steady state flowsheeting program, it has been specifically designed for acid gas treating applications. The program includes an extensive database of thermodynamic parameters, interaction coefficients, kinetics, etc. that has been optimized to accurately predict the vapor liquid equilibriums (VLE), thermodynamic and physical properties and the kinetically enhanced mass transfer (both analytical and rigorous) of amine based capturing processes. The program applies the Electrolyte Equation of State (E-EOS) thermodynamic model, which is expected to better predict the behavior of acid gas treating processes than conventional models often applied, like e.g. Kent-Eisenberg or more complex activity based models like Pitzer, Deshmukh-Mather or ElecNRTL. Alternative thermodynamic models can, however, easily be implemented in the simulator.

For the optimal prediction of column performances, the program includes a database of various tray types, as well as a large collection of both dumped and structured packing respectively. Several mass transfer and hydrodynamic models have been implemented that benefit from accurate physical property models (density, viscosity, surface tension, diffusivity, conductivity) specifically selected for acid gas treating applications.

The tool is able to describe complete acid gas treating processes, including complex processes with multiple (mixed or hybrid) solvent loops, and is able to significantly improve the understanding of the performance of potential new solvents.

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**Keywords:** flowsheeting; simulator; rate-based; gas treating; carbon dioxide capture; flue gas; absorption; stripping

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## 1. Introduction

Procede Gas Treating BV is involved in several projects aiming at the development of new cost-effective solvents which can be used in acid gas treating processes in general and carbon dioxide capture from power plant exhaust gas in particular. A simplified process scheme of a typical acid gas absorption process is shown in Figure 1.

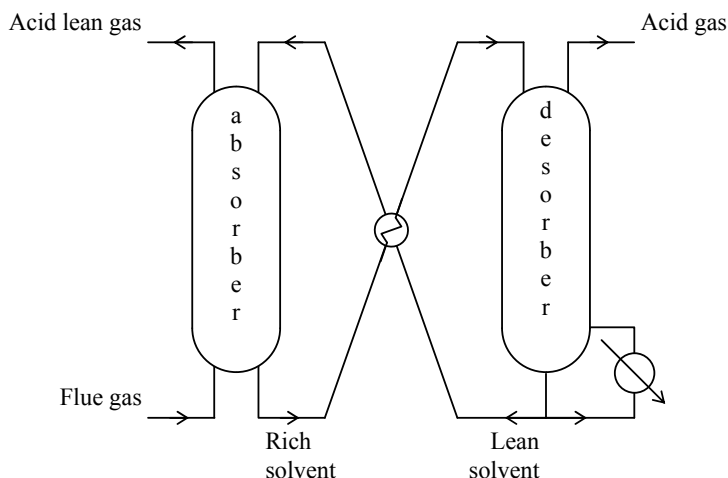


Figure 1: Simplified process scheme of an acid gas absorption process

In the absorber, the flue gas is contacted with absorption liquid. The acid gas ( $\text{CO}_2$ ) reacts with the absorption liquid and the cleaned flue gas is sent to the exhaust pipe. The loaded absorption liquid is heated and sent to the desorber where the absorption liquid is regenerated by desorption of the acid gases. The lean solvent is cooled down and can be re-used in the absorber.

For the development of these  $\text{CO}_2$  capture processes, detailed information about the thermodynamics, kinetics, physical properties and mass transfer phenomena is required. For optimization purposes, a detailed computer model that accurately describes the process is expected to be a useful tool. Such a computer model should enable the engineer to predict the performance of new solvent types (for example a new amine, a new blend of amines, a new hybrid solvent, etc.). The model also provides insight in the performance as a function of process parameters (for example temperature, pressure, column internals, etc.). Finally the tool should allow the engineer to predict the potential of different process layouts (for example side feeds and draws, addition of flash drums, heat integration, multiple solvent loops, etc.).

General purpose flowsheeting packages (like Aspen Plus<sup>®</sup>, Pro/II, Hysys<sup>®</sup>, etc.) are widely available on the market. These packages are however quite expensive, but more important these programs lack detailed knowledge of acid gas treating processes. Also, rate-based modeling of chemical absorption processes is often not supported at all or suffers from poor accuracy and numerical stability. Also, a limited number of specialized acid gas treating simulators / tools is available. Most of these programs are however academic tools which lack user-friendliness and are too complicated for the average end-user to apply.

To overcome the above mentioned limitations, it has been decided to develop a new dedicated acid gas treating simulator from scratch. Within the enterprise Procede Group BV, a lot of knowledge (mass transfer, thermodynamics, numerics) as well as a lot of basic coding (flash code, speciation code, mass transfer code, column models, numerical solvers, reporting) was already available. Combining the required expertise, recoding of existing models in a single developers platform and adding a powerful graphical user interface has resulted in a new acid gas treating simulator that – due to a better understanding of the processes – is expected to contribute to a significant performance improvement of existing as well as new acid gas treating processes.

## 2. Thermodynamic model (Electrolyte Equation of State)

The presently developed thermodynamic model is based on the Electrolyte Equation of State (E-EOS), as proposed by Fürst and Renon [1] and applied to acid gas treating by Solbraa [2], Derks et al. [3] and Huttenhuis et al. [4]. The basic equation for E-EOS is the Helmholtz free energy as a function of temperature, total volume and amount of molecules and ions in a specific phase. This equation gives a thermodynamic consistent system and all other thermodynamic properties origin from the partial derivatives of the Helmholtz energy. No additional equations or parameters are required. Since, we a cubic EoS is used, the equation is valid for the gas and liquid phase. The model should be able to model gas, liquid(s) and amine equilibrium (VLLE) in a consistent way. Extrapolation capabilities of the model are expected to be high. Prediction at high pressures, typically found in the natural gas industry, is expected to be more reliable compared to other thermodynamic models. The model can be extended to systems where different kind of inert components like hydrocarbons are present. The model reduces to a standard cubic equation of state if no ions are present in the solution, and by that enables consistent overall natural gas process simulation. The total Helmholtz free energy ( $A$ ) is a summation of several contributions:

$$A = A^{\text{IG}} + A^{\text{RF}} + A^{\text{SR1}} + A^{\text{SR2}} + A^{\text{LR}} + A^{\text{BORN}}$$

All contributions will be discussed separately. The ideal gas (IG) mixture of molecules and charged ions is the reference state of the E-EOS model. There are no interactions between the components in the ideal gas phase. Therefore, all derivatives to the amount of moles are not a function of the composition and the reference state of the model is independent on the composition of the mixture. This is an advantage when mixed solvents are used. The repulsive force term (RF) represents the change in Helmholtz free energy for the transfer of the neutral particles (molecules and ions) to the actual volume of the gas or the liquid phase and includes the excluded volume interactions. The two short range forces (SR1 and SR2) represent the change in Helmholtz free energy for the transfer of the neutral particles (molecules and ions) to the actual mixture and includes the short-range attractive dispersion forces. SR1 involves the term for the molecules and SR2 for the ions. The Huron Vidal [5] mixing rule is applied for the modeling of the interaction between the molecules. The long range ionic interaction term (LR) represents the change in Helmholtz free energy for the interaction between the ions in the mixture. These interactions are accounted for by the mean spherical approximation (MSA). The Born contribution (BORN) corresponds to the change in the Helmholtz free energy for discharging the ions in a vacuum (ideal gas) and recharging in a dielectric solvent, only taking into account the ion-solvent interactions at infinite dilution. This term has a major contribution to the Helmholtz free energy and is the reason for the low solubility of ions in the gas phase. The dielectric constant of the solvent mixture is the most important parameter in this term.

Partial derivatives of the total Helmholtz free energy give all thermodynamic properties. Since the Helmholtz free energy is a summation of different contributions, these contributions to the derivatives can be calculated separately. The first partial derivative to the total volume gives the total pressure of the system and can be used to calculate the density of the mixture as a function of temperature, pressure and composition. The first partial derivative to temperature gives the total absolute entropy of the mixture. Combination of the Helmholtz free energy and the entropy gives the internal energy and enthalpy of the mixture for the gas and liquid phase. Subsequently, it is possible to calculate the enthalpy of vaporization and enthalpy of reaction. The second partial derivative to temperature gives the isochoric heat capacity. With some additional derivatives this gives the isobaric heat capacity. The first partial derivative to the amount of moles for every component gives the chemical potential for every individual component in the mixture. Numerous properties originate from this derivative. The chemical potential, equilibrium constant of reaction, vapour liquid equilibrium factor, solubility factor (gas, liquid and solid), osmotic coefficient, vapour pressure, fugacity coefficient, activity coefficient, boiling point elevation and freezing point decrease are all based on the same derivative. So, large amount of different measurement are used to validate all the different partial derivatives of E-EOS, to get a thermodynamic consistent model.

### 3. Thermodynamic and physical properties

An overview of the main properties calculated for each stream in the flowsheet as well as all intersegment flows inside columns is shown in Table 1. These properties are calculated from models taken from various literature sources as discussed below.

Table 1 Thermodynamic and physical properties of streams

Property	Liquid phase	Vapor phase	Mixed phase
Enthalpy	Yes	Yes	Yes
Heat capacity	Yes	Yes	Yes
Density	Yes	Yes	Yes
Viscosity	Yes	Yes	No
Thermal conductivity	Yes	Yes	No
Surface tension	Yes	No	No

Enthalpy, heat capacity and density: These thermodynamic properties are directly derived from the thermodynamic model.

Vapor viscosity: Calculated by Lucas [6,7] method, which is based on a corresponding state method.

Liquid viscosity: Calculated from a modified implementation of the Teja-Rice equation [8,9]. The Teja-Rice method, which is based on a corresponding state method is suggested by Polling et al. [10] for aqueous solutions. The accuracy of the standard implementation of this method is, however, poor when applied to a system of more than two solvent species. The modifications made include the selection of two suitable reference fluids (water and a virtual amine), replacing the acentric factor by an adjustable pure component fit parameter and addition of an electrolyte correction [11,12] to adjust for the acid gas loading. The model does not require binary parameters. The model has been validated for seven different binary systems over a wide spread in temperature and concentration and was found to be very accurate. Typical errors are less than 5% which is very good for mixture viscosities that do not have binary fit parameters (Table 2). The maximum errors found are 25%, which is still fairly good, but it should also be noted that these values are not of any practical importance since they occur at conditions of equal molar fractions of amine and water, while practical solvents have significantly lower amine fractions (20% or less).

Table 2 Validation of liquid viscosity model

System	Independent sources	Total datapoints	Avg. error (%)	Max. error (%)
H2O –MDEA	12	249	3.8	21.6
H2O-MEA	5	51	-0.4	14.7
H2O-DEA	6	125	-1.7	25.2
H2O-TEA	4	51	-1.0	15.4
H2O-DGA	1	65	-0.7	19.1
H2O-DIPA	3	127	0.4	26.5
H2O-PPZ	4	79	-0.9	11.4

The expected quality for ternary mixtures is similar because the model is based on a virtual amine as a reference fluid. Unfortunately, there is only a small amount of data [13] of poor quality available for the viscosity of loaded solvents. Therefore it has not yet been possible to accurately qualify the accuracy of the implemented electrolyte adjustments, but for amines the accuracy is expected to be within 25% for loadings of up to 0.5 mol acid gas per mol amine.

Vapor thermal conductivity: Calculated from the Wassiliewa [14] equation. Pure component thermal conductivity is obtained from the method of Chung [15,16]. Binary interaction is found from the Mason and Saxena [17] modification.

Liquid thermal conductivity: Calculated from a modified version of the power law method. The standard power law method is not suitable for aqueous solutions or solutions that have a significant difference in the individual, pure component conductivities [10]. The Filippov [18] equation is believed to be very accurate for binary mixtures, but this method is not applicable to multi-component systems. The modification of the power law method applied in this model involves refitting of the fixed exponential value to match the Filippov equation as good as possible. In addition to this, an electrolyte adjustment has been added to the model to correct for the influence of acid gas loadings. Pure component thermal conductivities are required as input to the model and have to be taken from literature or to be measured.

Surface tension: Hadden [19] suggested a mol fraction based average. Polling [10] suggests a non-linear mole fraction based averaging method. However in the present study it was found that for amine systems, better results are obtained using a non-linear weight fraction based averaging method. At this moment an electrolyte adjustment has not been implemented yet. Pure component surface tensions are required as input to the model and have to be taken from literature or to be measured.

#### 4. Equilibrium-based unit operations

The simulator supports 17 types of equilibrium-based unit operations (Table 3).

Table 3 Supported equilibrium-based unit operations

Separation equipment	Separator, Flash drum, Column
Heat exchangers	Heater/cooler, Heat exchanger
Pressure manipulators	Pump, Turbine, Compressor, Expander, Appendage
Flowsheeting unit operations	Inlet, Outlet, Recycle, Mixer, Splitter
Gas treating equipment	Formulator, Thermodynamic model fitting

Basically all of these unit operations perform a flash calculation, including liquid phase speciation. The type of flash calculation performed depends on the specification of the unit operation. The simulator currently supports 6 types of flash calculations: PT, PQ, TQ, PV, TV, VQ (where PTQV stand for respectively Pressure, Temperature, Duty (or Enthalpy) and Vapor fraction).

The two phase gas-liquid isothermal equilibrium flash model (PT-flash) [20] is capable of calculating any point on the flash curve between the bubble point and the dew point. Given the feed composition  $z$  and the flash pressure and temperature ( $P$  and  $T$ ), an iterative calculation is used to find the vapor fraction ( $V$ ), vapor ( $y$ ) and liquid ( $x$ ) phase composition. The flash model solves the Rachford-Rice [21] equation, which is obtained by combining the component mole balances with the total mole balance and the phase distribution relation for 1 mole:

$$\text{Total mole balance for 1 mol: } F = L + V = 1$$

$$\text{Component mole balances: } z_i F = x_i L + y_i V$$

$$\text{Phase distribution relation: } K_i = y_i / x_i \quad (K_i \text{ is obtained from the thermodynamic model (Section 2)})$$

$$\text{Knowing that } \sum x_i - \sum y_i = 0, \text{ we can derive the Rachford-Rice equation: } f(V) = \sum_i \frac{z_i(1 - K_i)}{1 + V(K_i - 1)} = 0$$

The other flash algorithms use iterative PT-flash calculations to converge Q and/or V.

During each PT-flash iteration the liquid phase speciation is converged by applying a recoded and generalized version of the algorithm developed by Versteeg [22,23]. A simplified list of typical reactions is shown in Table 4.

Table 4 Typical reactions for amine based gas treating processes

Water dissociation	$2 \text{H}_2\text{O} \rightleftharpoons \text{H}_3\text{O}^+ + \text{OH}^-$
Bicarbonate formation	$2 \text{H}_2\text{O} + \text{CO}_2 \rightleftharpoons \text{H}_3\text{O}^+ + \text{HCO}_3^-$
Carbonate formation	$\text{H}_2\text{O} + \text{HCO}_3^- \rightleftharpoons \text{H}_3\text{O}^+ + \text{CO}_3^{2-}$
$\text{H}_2\text{S}$ dissociation	$\text{H}_2\text{O} + \text{H}_2\text{S} \rightleftharpoons \text{H}_3\text{O}^+ + \text{HS}^-$
Amine protonation	$\text{H}_2\text{O} + \text{R}_3\text{NH}^+ \rightleftharpoons \text{H}_3\text{O}^+ + \text{R}_3\text{N}$
Carbamate reversion (prim/sec-amine)	$\text{R}_3\text{NCOO}^- + \text{H}_2\text{O} \rightleftharpoons \text{R}_3\text{N} + \text{HCO}_3^-$

The main change to the work of Versteeg is that we have added non-ideality by introducing activity coefficients to the reaction equilibrium equation, for example the hydrolysis of  $\text{CO}_2$ :

$$K_R = \frac{x_{\text{true}, \text{H}_3\text{O}^+} \gamma_{\text{H}_3\text{O}^+} x_{\text{true}, \text{HCO}_3^-} \gamma_{\text{HCO}_3^-}}{(x_{\text{true}, \text{H}_2\text{O}} \gamma_{\text{H}_2\text{O}})^2 x_{\text{true}, \text{CO}_2} \gamma_{\text{CO}_2}}$$

Alternatively it is considered to implement an even more efficient algorithm that solves the speciation and flash equations simultaneously.

## 5. Hybrid unit operations: Columns (equilibrium-based and rate-based)

Columns have been implemented as hybrid unit operations, supporting both equilibrium- and rate-based calculations. A column performs an iterative segment-to-segment calculation. An equilibrium-based segment is essentially a QP-flash (usually adiabatic). The simulator currently supports 6 tray types, 6 main groups of dumped and 6 main groups of structured packing, all in different sizes and materials.

This section will focus on the rate-based segment implementation. A rate-based segment model has to simultaneously solve for the heat and the mass balances, including the mass transfer between the vapor and the liquid phase, the VLE at the interface and the liquid phase bulk speciation. The present rate-based model (Figure 2) is based on a completely recoded and revised version of the work of Versteeg [22] and van Elk [24]. The revisions include: Addition of a heat balance and heat transfer; taking into account the non-ideality of the chemical equilibrium; accounting for the mass transfer of all species.

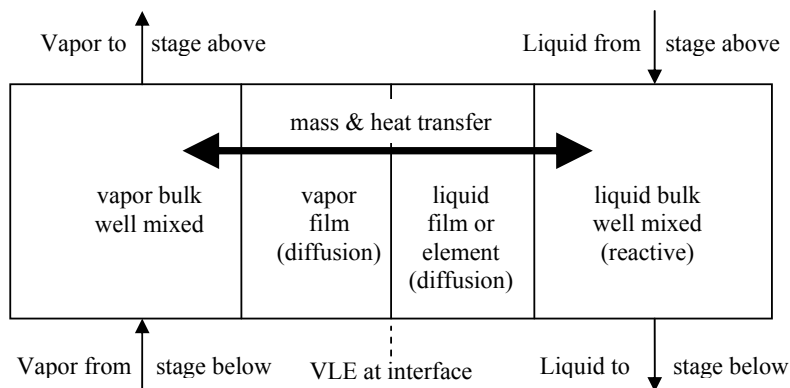


Figure 2: Schematic representation of the rate-based segment model (excluding side streams)

The correlations implemented in the simulator are discussed below.

Table 5 Supported hydrodynamic models (pressure drop and flooding)

Trays	Dumped packing	Structured packing
Perry [20]	Leva 1992 [25]	Bravo 1986 [26] Bravo 1992 [27]

Table 6 Supported mass transfer models ( $k_g$ ,  $k_l$ , area)

Trays	Dumped packing	Structured packing
Zuiderweg 1982 [28] Bennet 1993 [29] Scheffe 1987 [30] AIChE 1958 [31] Chan 1984 [32]	Onda 1968 [33] Bravo 1982 [34] Billet 1992 [35]	Bravo 1985 [36] Bravo 1992 [27] Shetty 1997 [37] Olujic (Delft model) 2002 [38] Onda 1968 [33] Billet 1992 [35]

Heat transfer parameters ( $h_g$ ,  $h_l$ ): The heat transfer parameters are estimated by application of the Chilton Colburn analogy that relates mass and heat transfer.

Diffusion coefficients ( $D_g$ ,  $D_l$ ): Vapor phase diffusion coefficients are calculated using the Fuller [39,40,41] algorithm, while Blanc's [42] law is used as a mixing rule. Liquid phase diffusion coefficients are calculated from a Wilke-Chang [43] method which has been slightly modified. CO<sub>2</sub> diffusion in amine solvents is based on the work of Versteeg [44].

Chemical enhancement of mass transfer: The simulator supports two different methods to deal with the chemical enhancement of mass transfer. The most accurate results are obtained by application of the rigorous method, which is based on the Higbie penetration theory [45]. This method applies a modified and recoded version of the work of Versteeg [22]. Alternatively, an analytical estimation method based on the work of Wellek [46] has been implemented. This method is less accurate, especially for mixed acid gas or mixed solvents, but requires much less computational efforts.

## 6. Conclusions / Future work

A new and user-friendly acid gas treating simulator has been developed. Future work to the simulator will, among others, focus on:

- Extending the pure component database (support more species).
- Extending the binary database (improve and extend the fitting of VLE).
- Fine tuning and further validation of physical property models.
- Add support for salts.
- Further improve thermodynamic consistency between speciation, VLE and enthalpy model.

## 7. Acknowledgement

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